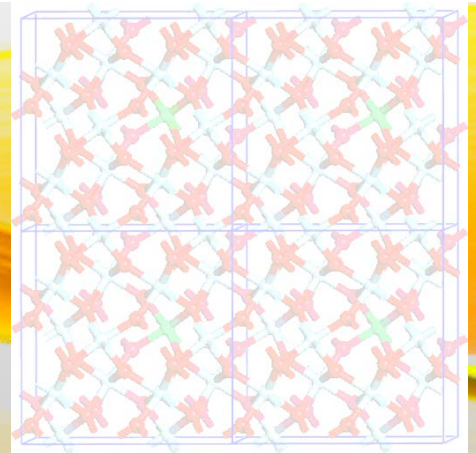
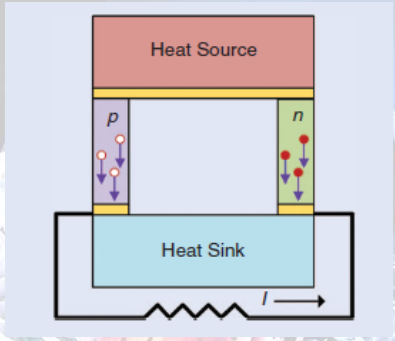
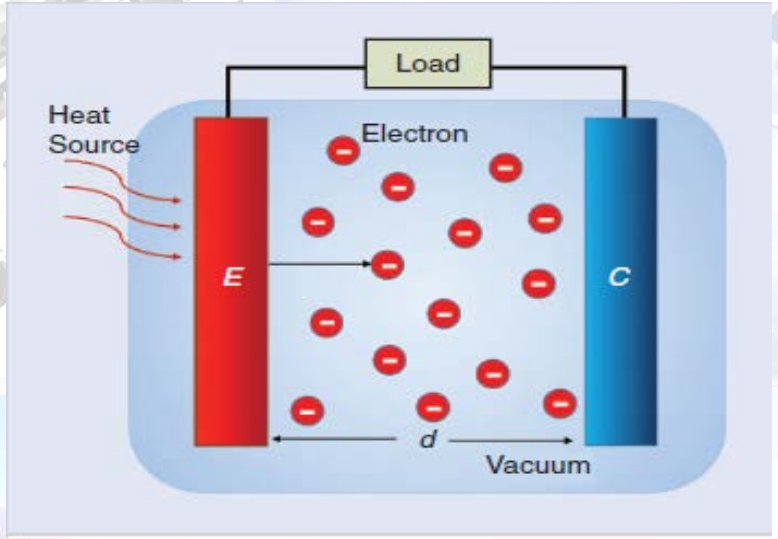
Ab initio Assessment of the Thermoelectric Performance of Ruthenium-doped Gadolinium Orthotantalate

Jon Goldsby, PhD , MBA
National Aeronautics and Space Administration
Glenn Research Center
Cleveland, Ohio



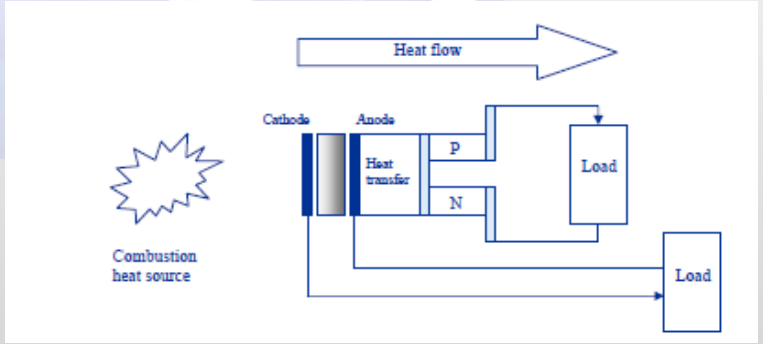
Concept Overview





$$J = AT^2 e^{-\left(\frac{E_w}{kT}\right)}$$

- Solid state energy harvesting using waste heat available in gas turbine engine offers potential for power generation to meet growing power needs of aircraft
- Thermoelectric material advances offer new opportunities
- Weight-optimized integrated turbine engine structure incorporating energy conversion devices

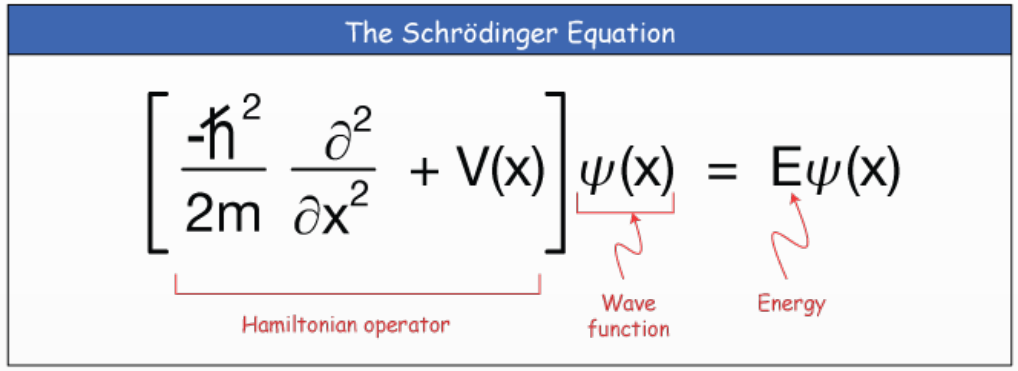


Thermionic
+
Thermoelectric

Characteristics for a desirable thermoelectric material

- Seebeck Coefficient ~ 100uV/K
- Electrical Resistivity 10^{-2} Ohm*cm
- Thermal Conductivity ~ 10 W/m*K
- Electronic Band Gap -must be greater than zero
- High Temperature Capability

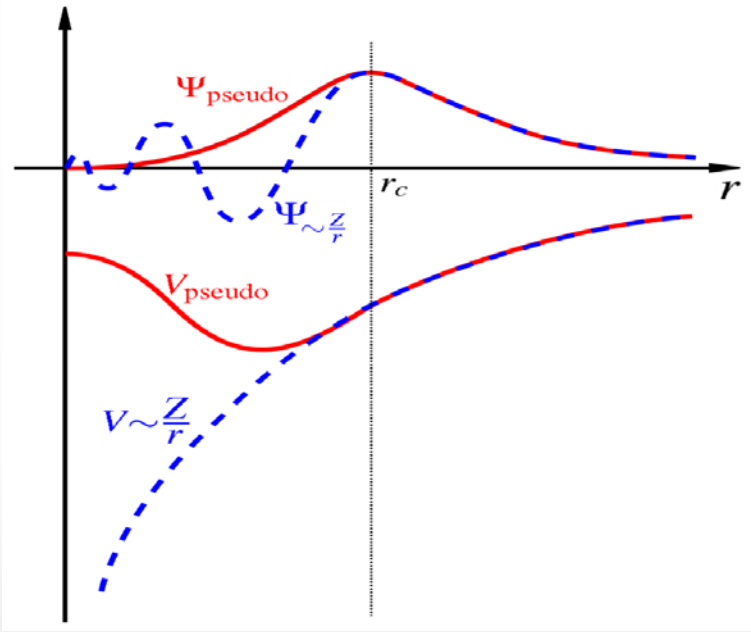
Computational Methods



BoltzTraP. A code for calculating band-structure dependent quantities ☆

Georg K.H. Madsen [a,*], David J. Singh [b]

Computer Physics Communications 175 (2006) 67–71

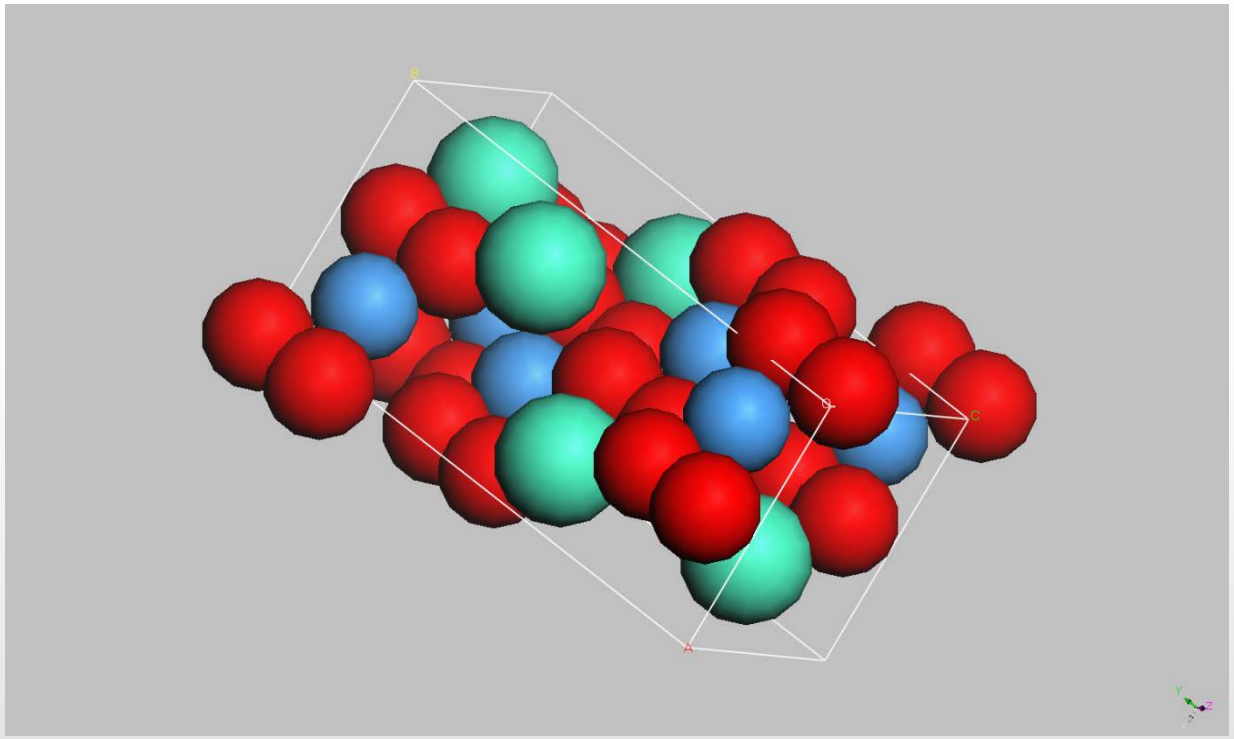


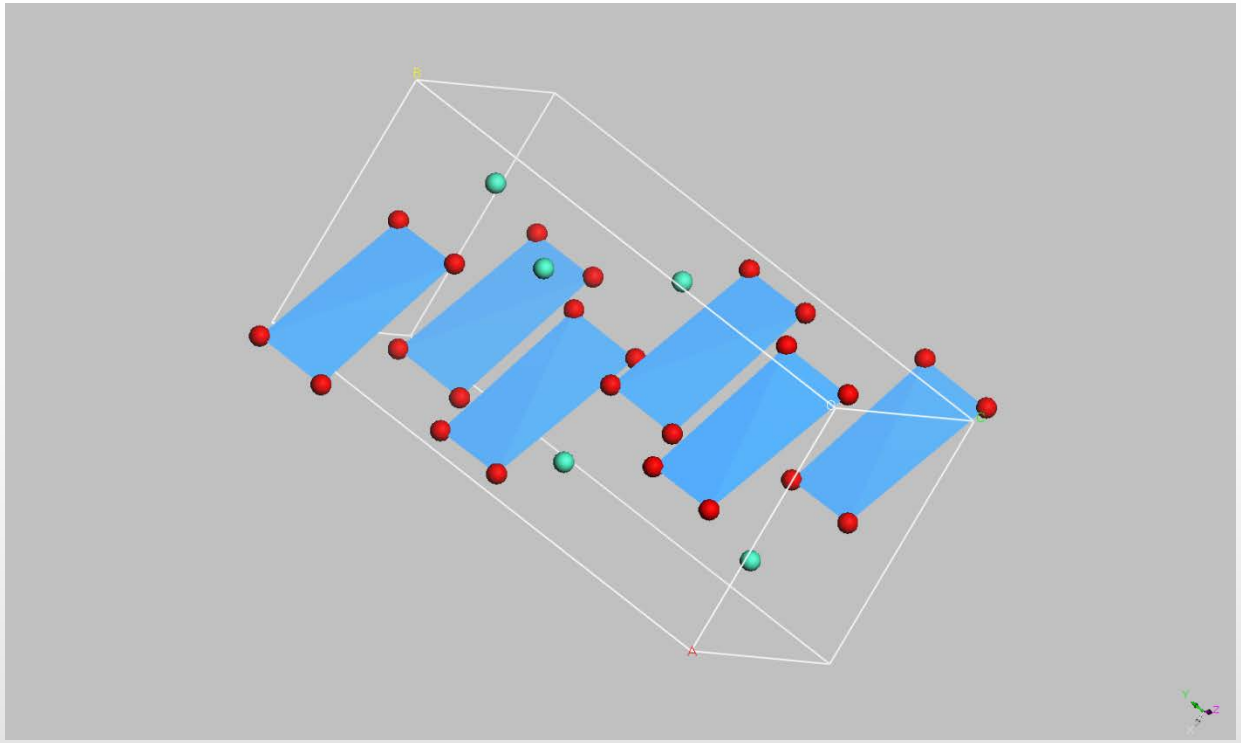
Vienna Ab-initio Simulation Package

The Vienna Ab-initio Simulation Package, better known as **VASP**, is a package for performing ab initio quantum mechanical molecular dynamics using either Vanderbilt pseudopotentials, or the projector augmented wave method, and a plane wave basis set

Computing Platform:
Hewlett-Packard Z840 Workstation
Dual 18-core intel Xeon processors
Widows 10 pro 64-bit
128 GB ram

$GdTaO_4$



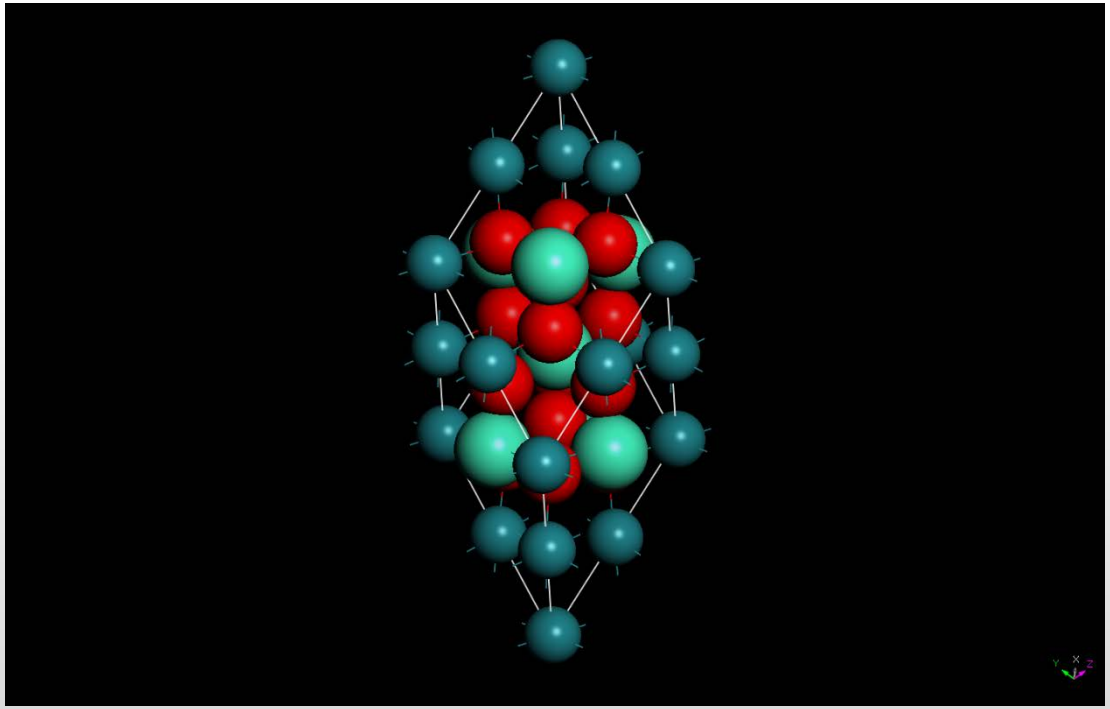


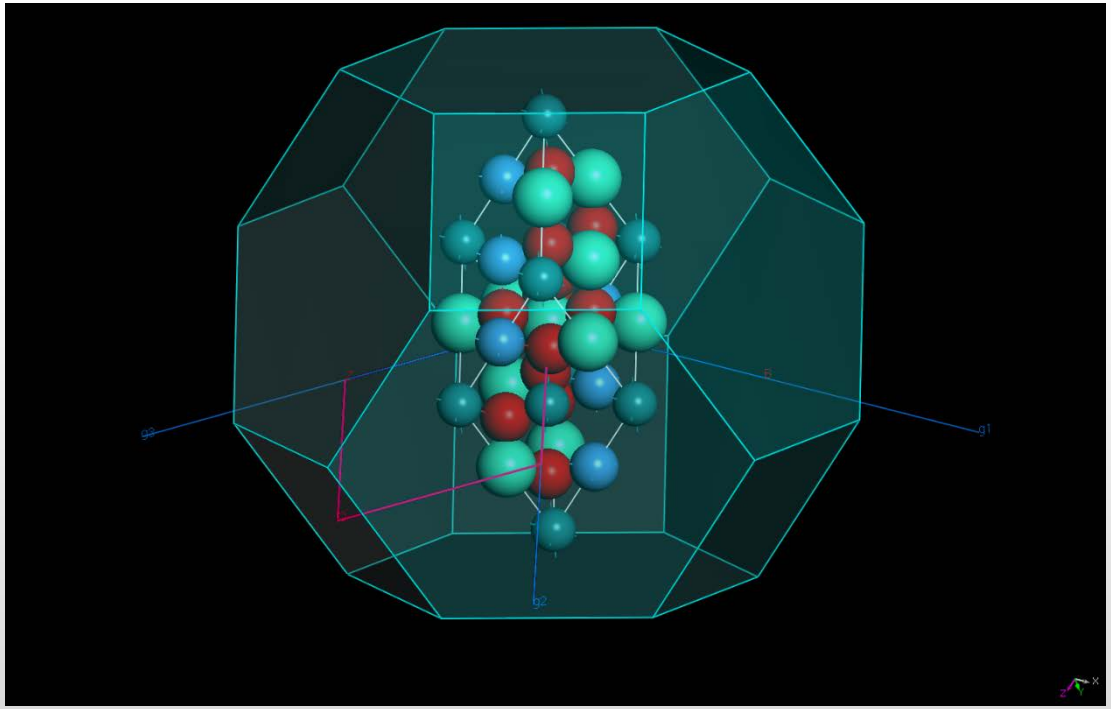
Gd –Green
Ta-Blue
O-Red

Complex Oxide – based Pyrochlores

mixed cation at B-site $A_2 (B^{3+}, B^{\sim 5+}) O_7$

Gd_2RuTaO_7

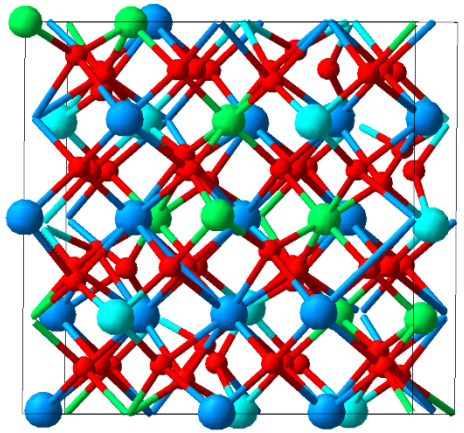




Calculated Cell Parameters

Parameter	Original	change	Final	%
a	10.091900	0.158505	10.250405	1.6
b	10.091900	0.137867	10.229767	1.4
c	10.091900	0.156757	10.248657	1.6
alpha	90.000000	-0.361354	89.638646	-0.4
beta	90.000000	0.392175	90.392175	0.4
gamma	90.000000	0.047839	90.047839	0.1
Volume	1027.824144	46.795542	1074.619686	4.6

Density: 8.759 Mg/m^3



Elastic constant matrix (GPa):

	1	2	3	4	5	6
1	303.99	122.74	112.40	0.00	0.00	0.00
2	122.74	342.62	128.66	0.00	0.00	0.00
3	112.40	128.66	289.89	0.00	0.00	0.00
4	0.00	0.00	0.00	98.03	0.00	0.00
5	0.00	0.00	0.00	0.00	76.22	0.00
6	0.00	0.00	0.00	0.00	0.00	88.62

Modulus	Voigt	Reuss	Hill
Bulk	184.90	183.56	184.23 GPa
Shear	90.75	89.78	90.27
Young's	233.98	231.59	232.78
Longitudinal			304.59

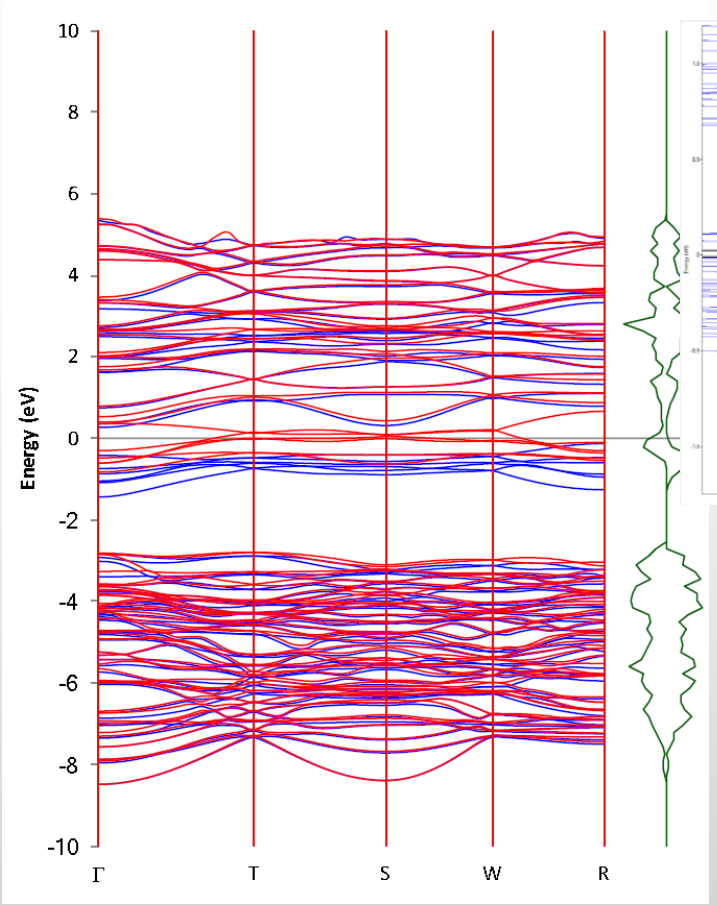
Velocity of sound

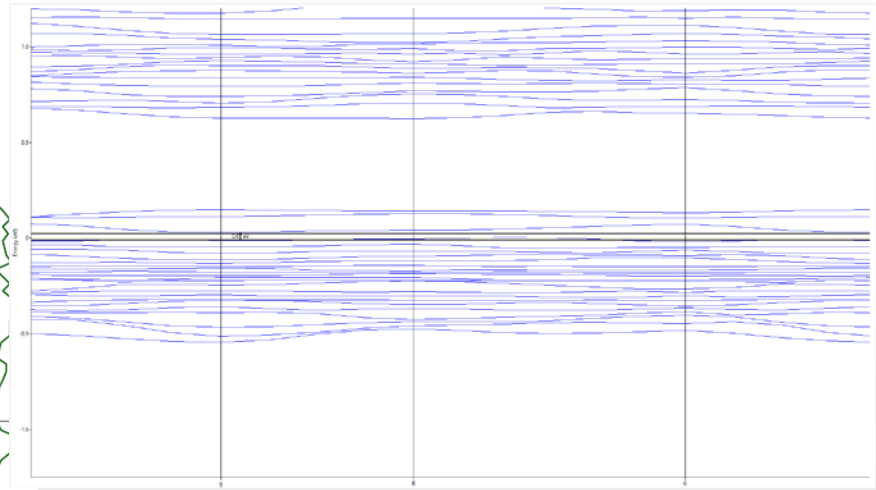
Calculated from Hill moduli:
transverse waves: 3271 m/s
longitudinal waves: 6009 m/s
mean: 3649 m/s

Debye temperature: 465.9 K
the thermal coefficient of linear expansion at 600K = 7.60×10^{-6}

Calculated Electronic Band Structure

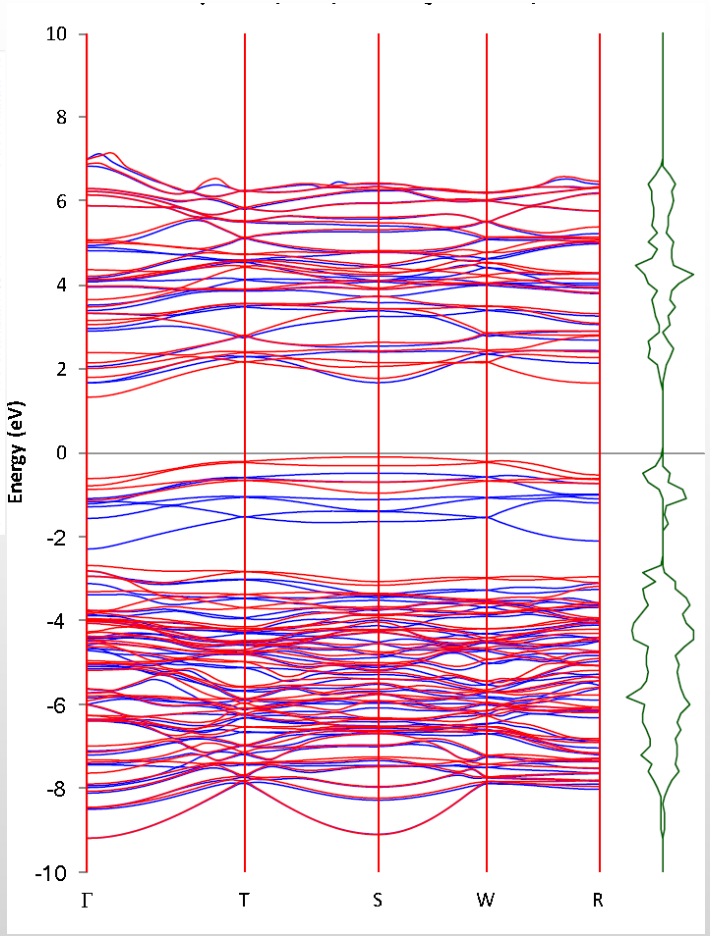
Perdew – Ernzerhof – Burke (PBE)



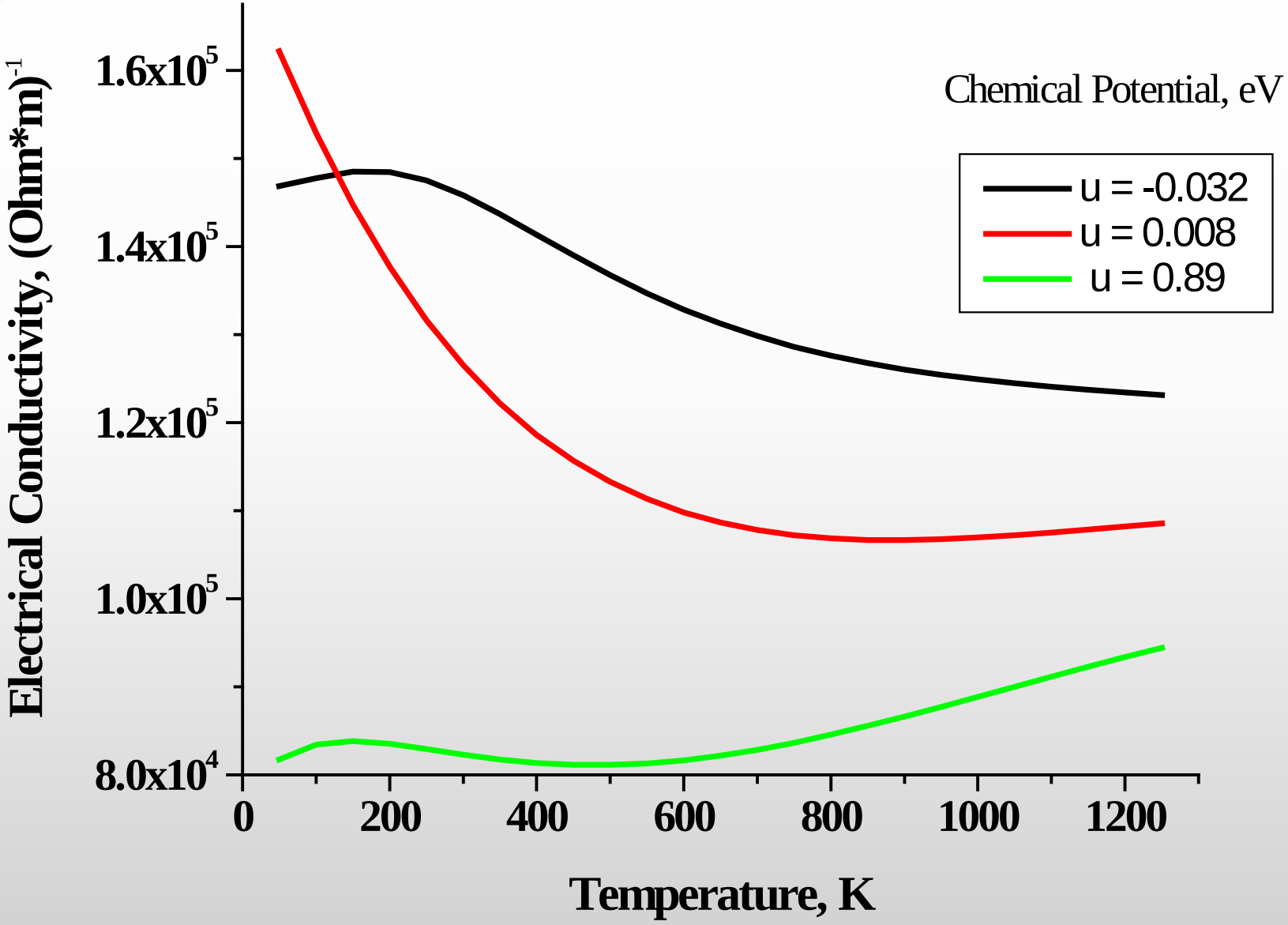


Cut off energy of 575 eV k-spacing 0.199/ A and 4x4x4 k mesh using DFT PBE the bandgap is 0.07 eV

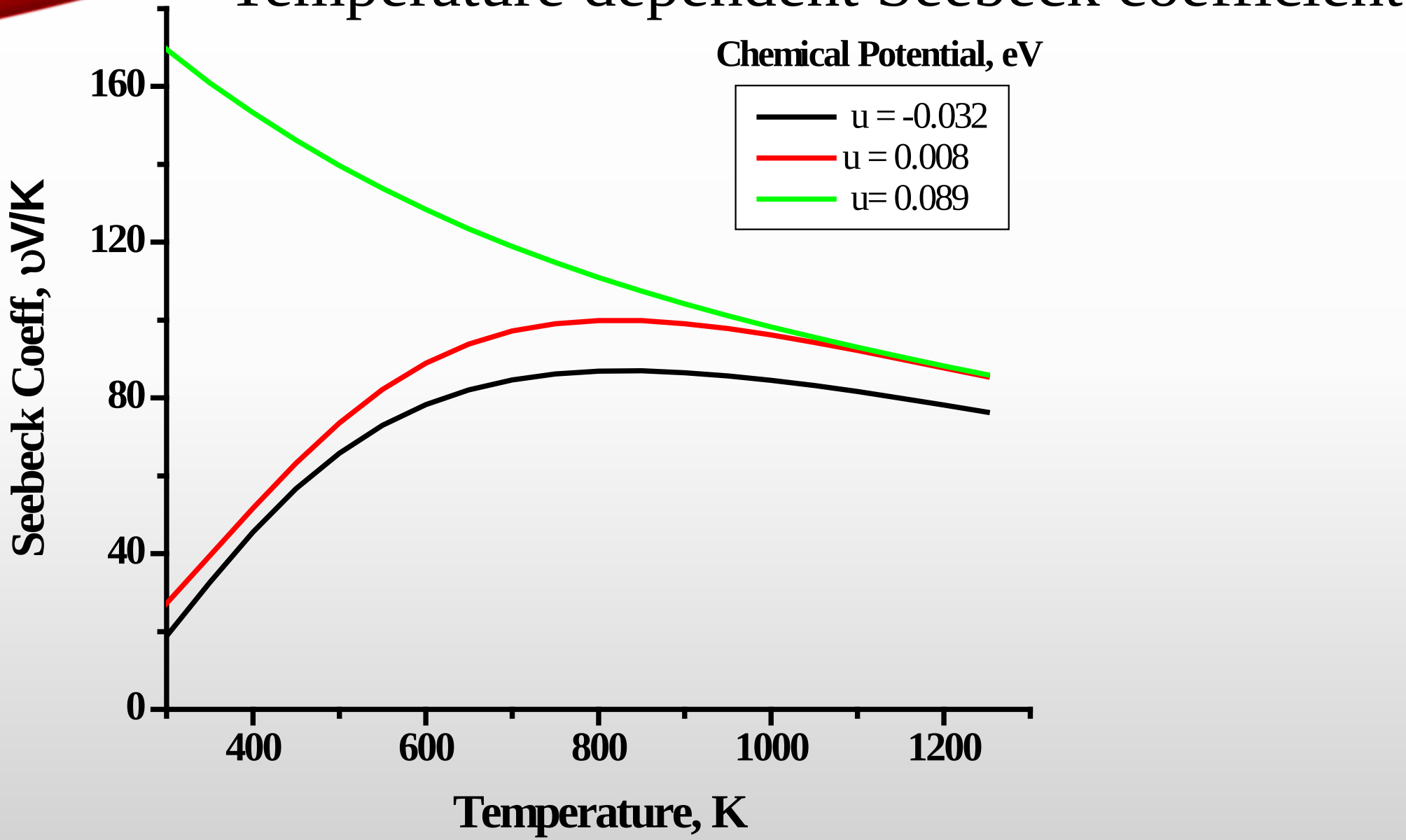
Heyd –Scuseria -Ernzerhof (HSE06)



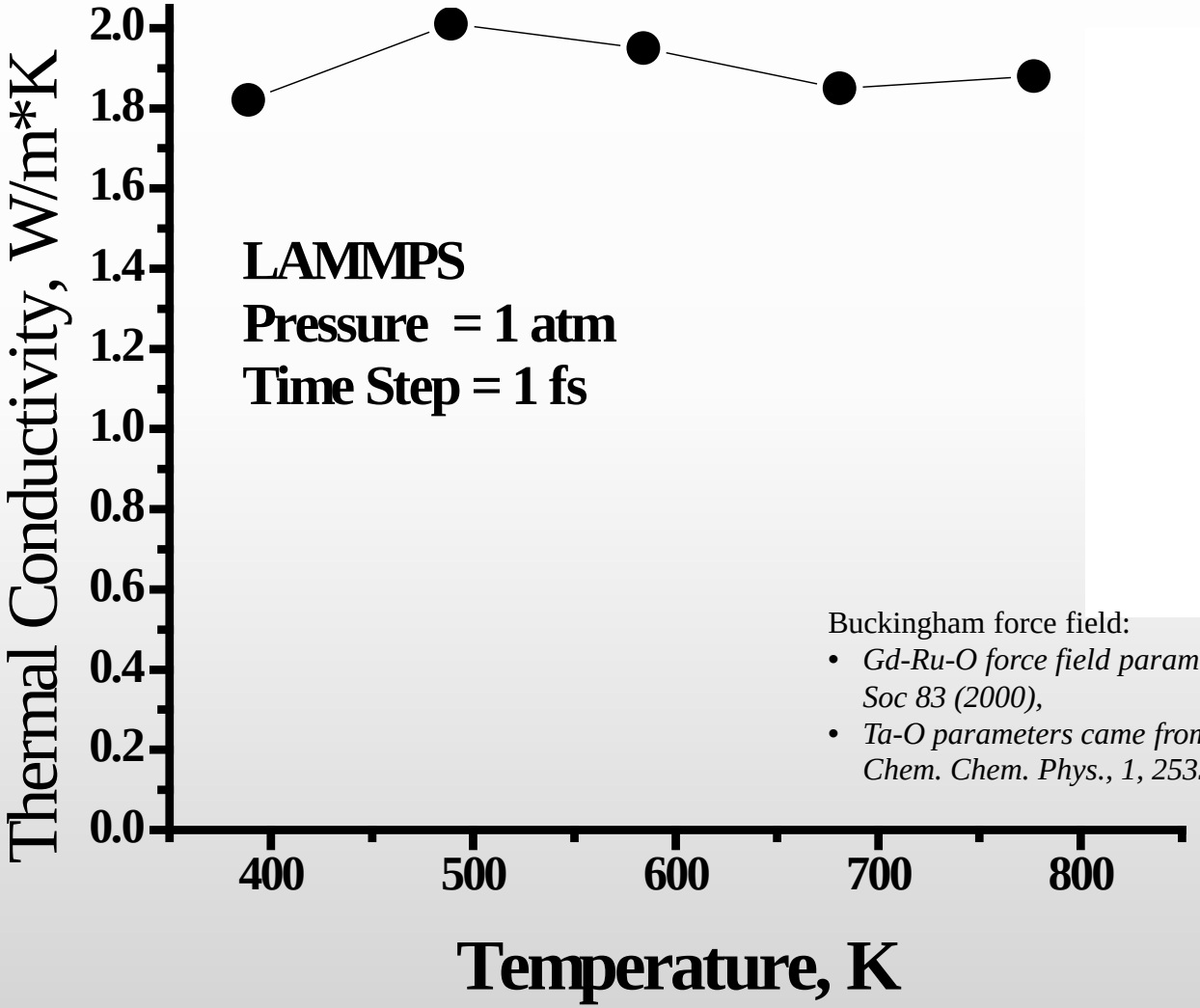
Electrical Conductivity

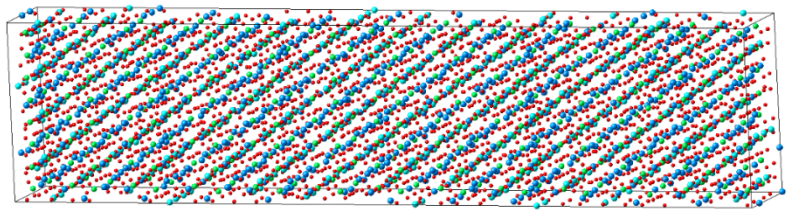


Temperature dependent Seebeck coefficient



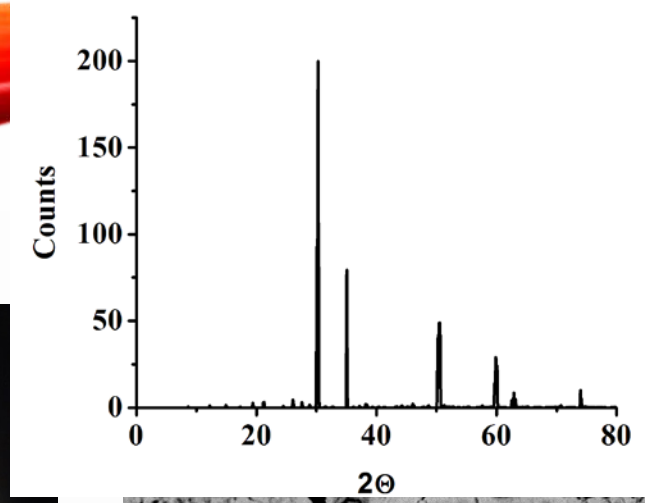
Molecular Dynamic Computational Results:
Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS)

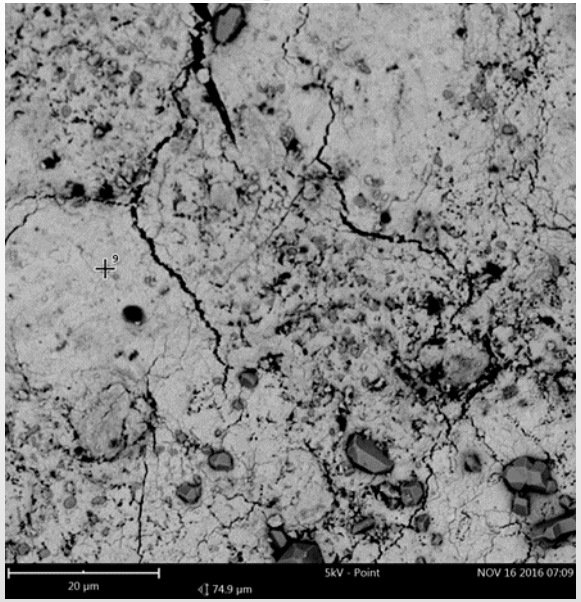


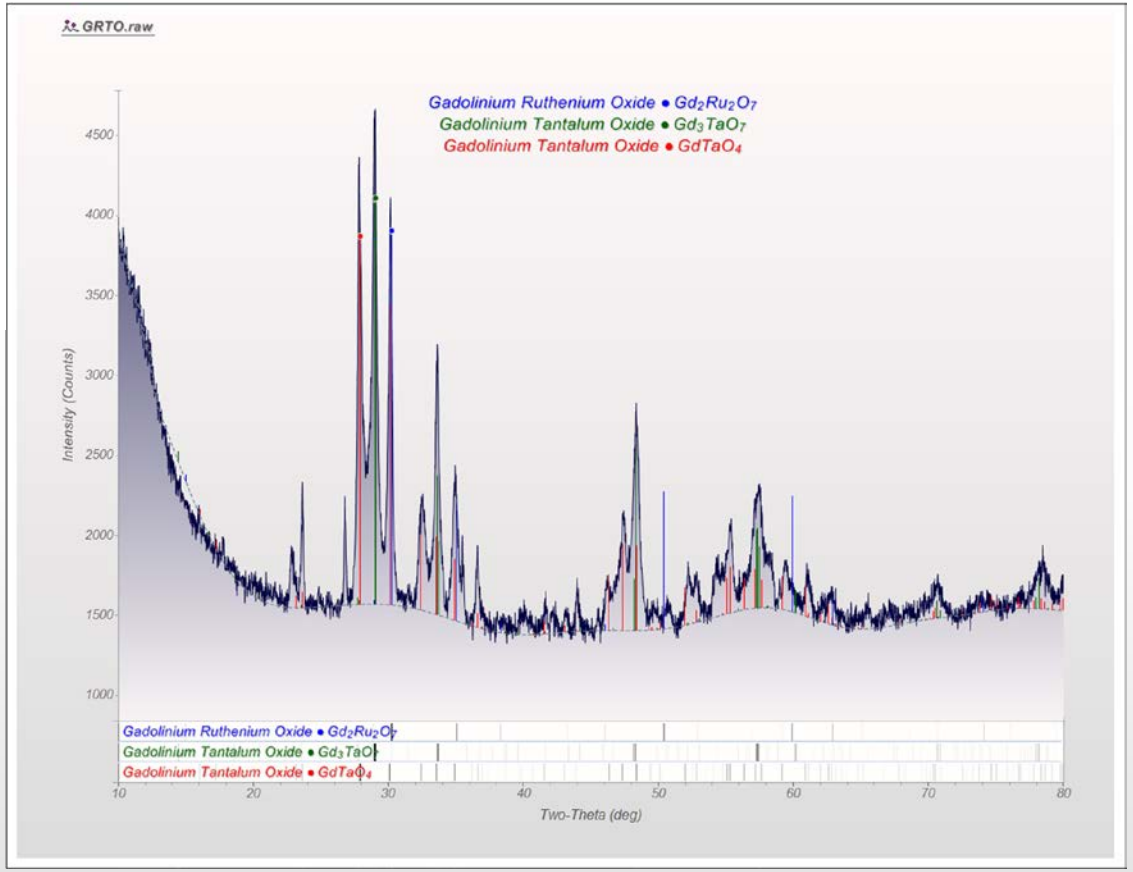


Buckingham force field:

- *Gd-Ru-O force field parameters came from Minervini, RW Grimes, KE Sickafus J Am Ceram Soc 83 (2000),*
- *Ta-O parameters came from S.M.Woodley, P.D.Battle, J.D.Gale and C.R.A.Catlow Phys. Chem. Chem. Phys., 1, 2535-2542 (1999).*







$$2Gd_2O_3 + Ta_2O_5 + 2RuO_2 \longrightarrow 2Gd_2(Ta, Ru)O_7 + \tfrac{1}{2}\,O_2$$

Solid state reaction, mechanical mixing, sintering in air, hot pressing

CONCLUSIONS

- Potential exist to harvest electrical power from excess enthalpy from gas turbine engines.

- Computational methods have enabled some fundamental parameters to be predicted in the development of thermoelectric materials.

- Some descriptions (band structure) are very sensitive to such things as mesh density.

- Oxide pyrochlores have potential as thermoelectric materials.